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Spin and Geometric Phase Control Four-Wave Mixing from Metasurfaces

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The spin angular momentum of light plays an important role in nonlinear interactions in optical systems with rotational symmetries. Here, the existence of the nonlinear geometric Berry phase is demonstrated in the four-wave mixing process and applied to spin-controlled nonlinear light generation from plasmonic metasurfaces. The polarization state of four-wave mixing from the ultrathin metasurfaces, comprising gold meta-atoms with four-fold rotational symmetry, can be controlled by manipulating the spin of the excitation beams. The mutual orientation of the meta-atoms in the metasurface influences the intensity of four-wave mixing via the geometric phase effects. These findings provide novel solutions for designing metasurfaces for spin-controlled nonlinear optical processes with inbuilt all-optical switching.

1. Introduction

Nonlinear optical processes strongly depend on both the constituent materials and the symmetries of an optical system [1, 2]. Recent advances in nonlinear optics have shown that the conventional selection rules of nonlinear processes in crystals can also be applied to plasmonic metasurfaces [3-6]. In general, the symmetry of a metasurface or metamaterial is determined by both the local symmetry of each individual meta-atom and the global symmetry of the lattice [7-20]. For example, the global symmetry of an assembly of centrosymmetric plasmonic meta-atoms can be broken by introducing passive elements into the metasurface lattice, activating second-harmonic generation (SHG) [8].

The presence of inversion symmetry in a material is mainly a consideration for even-order nonlinear optical processes such as SHG [6-20]. By contrast, the rotational symmetry of nonlinear optical crystals is important when considering both even- and odd-order harmonic generation [21, 22]. Under excitation with a fundamental wave carrying spin angular momentum (SAM), such as that carried by circularly polarized light, the SAM state of light generated through nonlinear interactions in a crystal with rotational symmetry can be determined according to certain selection rules [21-27]. For circularly polarized harmonic generation in the presence of m -fold rotational symmetry, the allowed n^{th} harmonic is $n = l(m \pm 1)$, where l is a positive integer, and the choice of positive or negative sign corresponds to harmonics generated with the same and opposite SAM, respectively, compared to that of the fundamental wave [21-27]. Recently, these selection rules, formulated for conventional optical crystals, have been successfully applied to nonlinear metasurfaces [14, 23-27]. For example, by designing the gold plasmonic metasurfaces with three- and four-fold rotational symmetries, only SHG and third-harmonic generation (THG) with opposite SAM state with respect to the fundamental wave are allowed [14, 23, 24].

Four-wave mixing (FWM) is a third-order nonlinear optical process which has important applications in optical parametric amplification, supercontinuum generation, optical

phase conjugation [28], optical switching and frequency comb generation [29]. In the field of plasmonics and metamaterials, electric-field-enhanced FWM has been extensively studied [30-36]. However, most these works focus on linearly polarized excitation light. The circular dichroism of FWM was observed in a bulk chiral metamaterial at microwave frequency, but only pump and signal waves with the same circular polarizations were discussed.

Here, we show that the phase and intensity of the nonlinear polarization of FWM from a metasurface with four-fold (C_4) rotational symmetry can be geometrically controlled through a spin coupling effect reliant on the geometric (Pancharatnam-Berry) phase. To the best of our knowledge, this is the first time the geometric phase and the spin-switching effect of FWM is experimentally demonstrated by controlling the polarization of the input beams. The geometric phase effects have been discussed for harmonic generation in which only the spin of one pump wave needs to be considered [24]. FWM, however, is a more complicated, multi-wavelength process, which provides more degrees of freedom in designing and controlling output signals, since it depends on multiple spin states of the input light and multiple plasmonic and material resonances of the system. FWM is more versatile and important for practical applications. Our results demonstrate that using circularly polarized light and exploiting the Berry phase leads to increased freedom in the design of nonlinear metasurfaces, with significant implications in the design of nonlinear optical devices for various functionalities.

2. Results and Discussion

We study FWM from the metasurfaces formed by square arrays of subwavelength gold nanocross meta-atoms (**Figure 1a,b**; see fabrication details in Supporting Information). The meta-atom geometry, exhibiting a plasmonic resonance at a wavelength of 1310 nm (**Figure 1c**), belongs to the C_{4h} point group, being symmetric under four-fold rotation and reflection about two axes (the meta-atoms lie in the xy -plane). We consider the semi-degenerate FWM

process at a frequency $\omega_{\text{FWM}} = 2\omega_1 - \omega_2$, where ω_1 and ω_2 are the frequencies of “pump” and “signal” waves, respectively. In this case, the nonlinear polarization can be expressed as[2]

$$\vec{P}^{\text{FWM}}(2\omega_1 - \omega_2) = \epsilon_0 \chi^{(3)} \vec{E}_{\omega_1} \vec{E}_{\omega_1} \vec{E}_{\omega_2}^* \quad , \quad (1)$$

where \vec{E}_{ω_1} and \vec{E}_{ω_2} are the electric fields of pump and signal waves, respectively, $\chi^{(3)}$ is the third-order nonlinear susceptibility tensor, and ϵ_0 is the vacuum permittivity. Assuming both the pump and signal waves are collinearly incident on the meta-atom along the z-axis, the effective nonlinear susceptibility tensor $\chi^{(3)}$ can be reduced to a few independent elements [2]: $\chi_1 = \text{xxxx} = \text{yyyy}$; $\chi_2 = \text{xxyy} = \text{yyxx} = \text{xyxy} = \text{yxyx}$; $\chi_3 = \text{xyyx} = \text{yxyx}$. For spin-carrying, circularly polarized light, the input fields can be expressed as $\vec{E}_{\omega_1} = a(\hat{e}_x \pm i\hat{e}_y)$ for the pump wave and $\vec{E}_{\omega_2} = b(\hat{e}_x \pm i\hat{e}_y)$ for the signal wave, where the ‘+’ and ‘-’ signs correspond to left-handed (LCP) and right-handed (RCP) circular polarizations, respectively, and a and b are the complex amplitudes. The same considerations can be applied to other FWM frequencies, such as $2\omega_2 - \omega_1$, $2\omega_1 + \omega_2$, and $2\omega_2 + \omega_1$.

Figure 1a summarizes the symmetry selection rules for the spin of FWM light generated from a meta-atom with C4 symmetry (see Supporting Information for derivations). We contract “LCP” as “L” and “RCP” as “R”, and label the states of the incident polarization as LLL, LRR, RLL, or RRR, where the first letter identifies the handedness of the signal wave ω_2 , the following two of the pump wave ω_1 . The selection rules are LLL-L, RRR-R, LRR-L and RLL-R, where the appended index indicates the spin of the FWM wave at ω_{FWM} . The polarization of the signal wave controls the spin state of the output FWM, which provides a basis for the switching and modulation of FWM by controlling the polarization of the input beams. For example, if the spin state of the signal wave is L, the nonlinear polarization at the FWM frequency is given by $\vec{P}^{\text{FWM}}(2\omega_1 - \omega_2) \propto a^2 b (\chi_1 \pm \chi_2 - \chi_3) (\hat{e}_x + i\hat{e}_y)$. If the spin

state of the signal wave is flipped to R, the FWM polarization also changes $\vec{P}^{\text{FWM}}(2\omega_1 - \omega_2) \propto a^2 b(\chi_1 \mp \chi_2 - \chi_3)(\hat{e}_x - i\hat{e}_y)$. Switching is passive in this case and relies on external control of input light polarization; an active, an active all-optical scheme may also be possible by exploiting the intensity-dependent refractive index and its influence on the polarization state of the input beams[37].

Experimentally, the FWM intensity and polarization from the Au nanocross metasurface was studied using a signal wave at the plasmonic resonance (1310 nm) and a pump wavelength at 1028 nm, giving FWM at 846 nm (see Supplementary Information for the details of optical measurements). Peak FWM conversion efficiency relative to the signal beam is estimated from the measurements to be of the order of 10^{-8} in our experimental conditions. The polarization analysis proves that the FWM wave is circularly polarized: it produces a sinusoidal trace with the rotation of the analyser placed after the quarter-wave-plate (**Figure 1d, I**). The FWM polarization trace shifts by $\pi/2$ if the signal wave polarization is switched, indicating a change in the handedness of the FWM light, as described by the selection rules above. Similar measurements performed in all polarization configurations for the pump and signal beams confirm that the handedness of the FWM is the same as that of the signal beam, in agreement with the theory and the numerical simulations (**Figure 1d, II**). The relative FWM intensity of the cross-polarized cases is higher in the simulations because of fabrication and measurement imperfections. As a control, a similar set of measurements was performed from the ITO-coated glass substrate next to the metasurface (**Figure 1d, III**). The ratios between the cross-polarized (LRR-L and RLL-R) and co-polarized (LLL-L and RRR-R) FWM on the metasurface and substrate are about 0.1 and 0.006, respectively. The much higher relative FWM emission from the array (cross-polarized FWM/co-polarized FWM) can be understood taking into account the selection rules since the FWM is not generated in the substrate in the cross-polarized case (see Supporting Information), which is also confirmed in

the simulations (**Figure 1d, IV**). A consequence of the selection rules is that only the metasurface contributes to FWM in the cross-polarized cases, while in the co-polarized cases the substrate contributes as well.

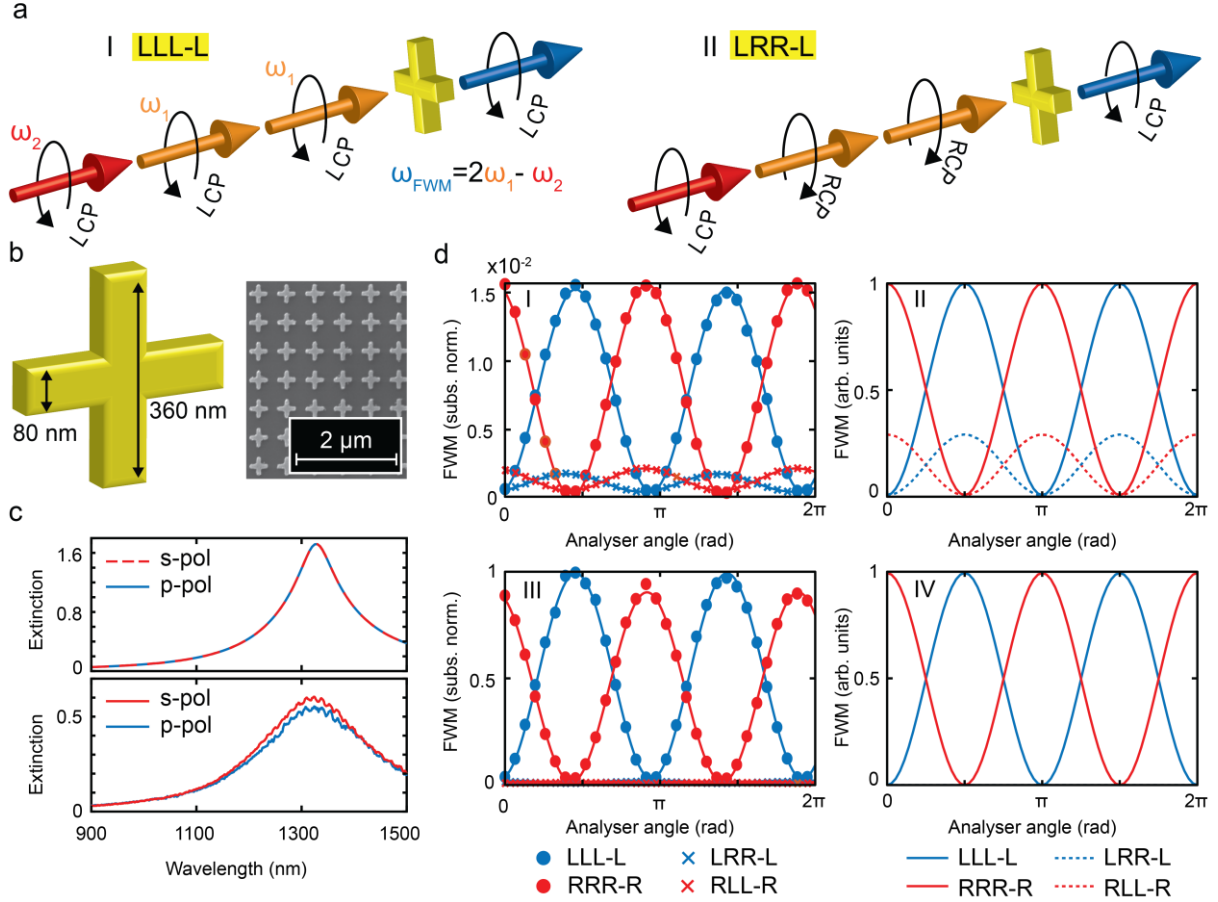


Figure 1. (a) Illustration of the FWM (frequency ω_{FWM}) selection rules for the co-polarized LLL-L (I) and cross-polarized LRR-L (II) signal (frequency ω_2) and pump (frequency ω_1) beams. The complete set of the selection rules is presented in Supporting Information. (b) Schematic of a meta-atom with C4 rotational symmetry and an SEM image of the metasurface. (c) Theoretical (top) and experimental (bottom) extinction spectra of the metasurface at normal incidence for different orientations of linearly polarized light. (d) Polarization analysis of the FWM signal from the metasurface (I, II) and the substrate (III, IV) performed by converting the circular polarization of the FWM to linear with a quarter-waveplate and rotating an analyser: (I, III) experimental data (symbols) are fitted with $\sin^2\phi$ curves (solid lines) and (II, IV) simulations. Experimental data are normalised to the substrate FWM. A change of the handedness of circularly polarized FWM is observed as a $\pi/2$ shift between the traces.

If the meta-atom has a relative orientation angle of θ with respect to the laboratory reference system (Figure 2), and both the pump and signal waves have the same spin state σ

(circular polarization of the same handedness), they both acquire a geometric Berry phase factor of $e^{i\sigma\theta}$ in the local coordinate system of the meta-atom [5]. The local nonlinear polarization in this frame of reference is given by $\vec{P}_{\text{loc}}^{\text{FWM}}(2\omega_1 - \omega_2) \propto e^{i\sigma\theta} a^2 b(\chi_1 \pm \chi_2 - \chi_3)$. After transforming back to the laboratory coordinates, we have $\vec{P}_{\sigma}^{\text{FWM}}(2\omega_1 - \omega_2) \propto a^2 b(\chi_1 + \chi_2 - \chi_3)$. Thus, the phase of the nonlinear polarization is independent of the orientation angle of the nanocross. On the other hand, if the spin state of signal wave is opposite ($-\sigma$) from that of the pump wave (σ), it experiences a geometric phase of $e^{-i\sigma\theta}$, and the resulting nonlinear polarizability of the FWM is $\vec{P}_{-\sigma}^{\text{FWM}}(2\omega_1 - \omega_2) \propto a^2 b e^{i4\sigma\theta}(\chi_1 - \chi_2 - \chi_3)$ in the laboratory system of coordinates. Thus, the nonlinear geometric Berry phase with values of 0 and $4\sigma\theta$ is introduced into the FWM signal for pump and signal waves with the same (LLL-L, RRR-R) or opposite (LRR-L, RLL-R) spin states, respectively.

To experimentally prove the presence of the nonlinear geometric Berry phase in FWM processes, a set of metasurfaces was fabricated with different orientations of the meta-atoms. In each unit cell of these metasurfaces, there are two gold meta-atoms (C4 symmetry) with different orientation angle $\pm\theta$ with respect to the y axis (**Figure 2a**). Under the LLL-L and RRR-R polarization configurations, the relative geometric phase introduced in the electric field of the FWM signals from the two meta-atoms in each unit cell equals zero, so the far-field FWM intensity is formed by constructive interference from the meta-atoms and similar far-field intensity is measured for all studied metasurfaces. Conversely, the geometric phase of the electric field of the FWM signals equals $\pm 4\sigma\theta$ for the two meta-atoms under the RLL-R and LRR-L polarization configurations, and the intensity of FWM from each unit cell is $I(2\omega_1 - \omega_2) \propto |e^{i4\sigma\theta} + e^{-i4\sigma\theta}|^2 \propto \cos^2(4\theta)$. If $\theta = 0^\circ$ or $\theta = 22.5^\circ$, the FWM from the two meta-atoms interfere in the far-field constructively and destructively, respectively. In all cases, FWM is locally generated in the near field of the individual nanocrosses, but for some

nanocross orientations destructive interference takes place in the far field, causing part of the FWM to be dissipated due to Ohmic losses in the nanostructures.

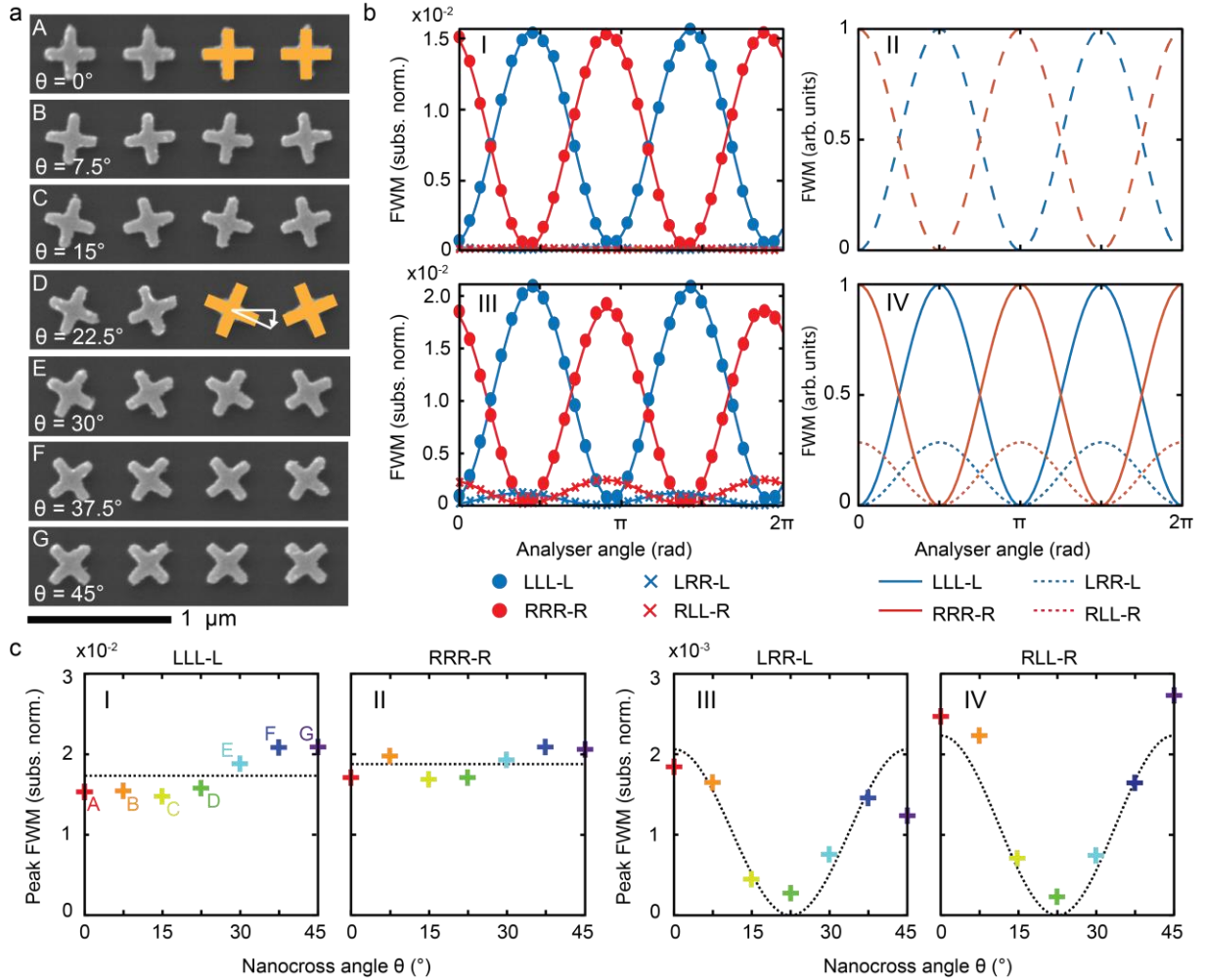


Figure 2. (a) SEM images of the two consecutive unit cells of the metasurfaces A—G with different meta-atom rotation angles θ indicated in each image. (b) FWM polarization trace (as in Fig. 1d) for metasurfaces D ($\theta = 22.5^\circ$; I, II) and G ($\theta = 45^\circ$; III, IV): (I, III) experimental data (symbols) are fitted with $\sin^2\phi$ dependences (solid lines) and (II, IV) simulations. A change of the handedness of circularly polarized FWM is observed as a $\pi/2$ shift between the traces. (c) FWM intensity from metasurfaces A—G for co-polarized (I, II) and cross-polarized (III, IV) signal and pump beams. The plots in I, III and II, IV are normalised to the intensity of the FWM signal from the substrate for the LLL-L and RRR-R cases, respectively. Experimental data (symbols) corresponds to the maxima of the $\sin^2\phi$ fits. Simulations (dashed lines) are normalised to the mean experimental signals.

FWM measurements and calculations for all the metasurfaces were compared for all polarization combinations (examples in **Figure 2b**, see Supporting Information for the

complete data set). The FWM intensities are extracted from each set of measurements (**Figure 2c**) show that for the LLL-L and RRR-R polarization configurations, the experimental FWM intensities from the metasurfaces do not depend on the orientation angle of the meta-atoms, as predicted by the theory (slight variations are probably due to imperfections in fabrication or the change in interaction strength between meta-atoms as they are rotated in the unit cell). For the LRR-L and RLL-R polarization configurations (**Figure 2c** III, IV), there is strong modulation of the FWM intensity with the rotation angle with $\cos^2(4\theta)$ dependence.

In order to better understand the spin-selective and nonlinear geometric Berry phase of FWM, we calculated the spatial distribution of nonlinear FWM polarization on the metasurfaces with $\theta = 0^\circ, 22.5^\circ, 45^\circ$ (**Figure 3**). The amplitude distributions of $\vec{P}_{\text{LRR-L}}^{\text{FWM}}(2\omega_1 - \omega_2)$ and $\vec{P}_{\text{RLL-R}}^{\text{FWM}}(2\omega_1 - \omega_2)$ (**Figure 3a**) show strong field enhancement at the edge of the meta-atoms, due to the localized plasmonic resonance of the signal wave at a wavelength of around 1310 nm. The effect of rotation is evident in the phase distributions of the nonlinear polarizations for both LRR-L and RLL-R polarization, configurations exhibiting 4-fold rotational symmetry, which can be described by a topological charge of $q = \pm 4$ [38] (**Figure 3b**). This is not observed in the co-polarized cases LLL-L and RRR-R, for which the phase distributions do not show the characteristic four-fold rotational symmetry (see Supporting Information). After adding $\pm 4\theta$ to the phases of $\vec{P}_{\text{LRR-L}}^{\text{FWM}}(2\omega_1 - \omega_2)$ and $\vec{P}_{\text{RLL-R}}^{\text{FWM}}(2\omega_1 - \omega_2)$, respectively, the same phase distributions are observed on the three metasurfaces in their local coordinates (**Figure 3c**), confirming the existence of the nonlinear geometric Berry phase $\pm 4\sigma\theta$ in the nonlinear polarization of FWM.

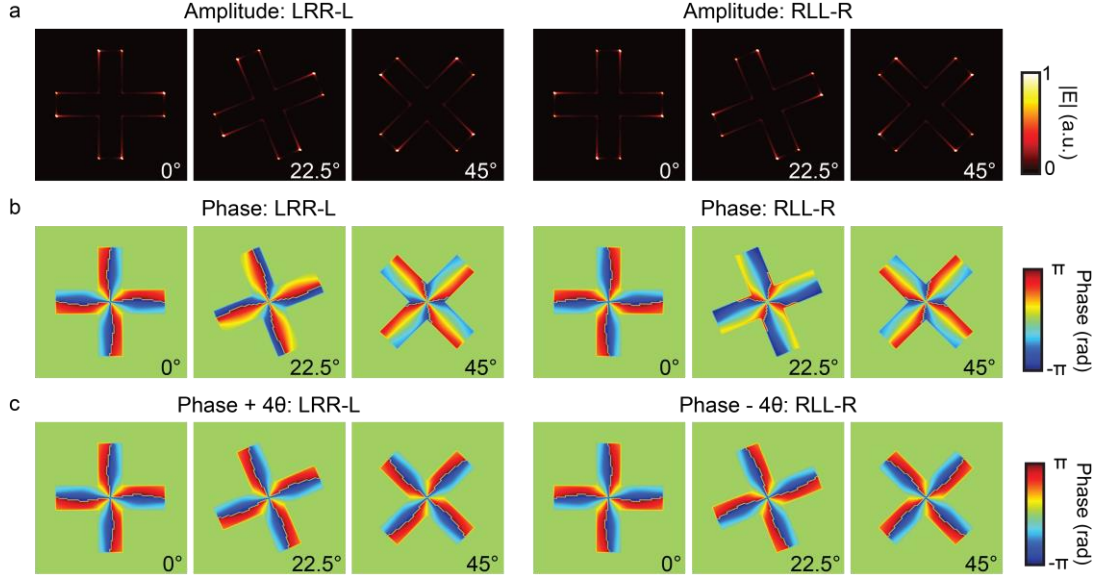


Figure 3. (a) Amplitude and (b) phase distributions of the nonlinear polarization of FWM $\vec{P}^{\text{FWM}}(2\omega_1 - \omega_2)$ on metasurfaces A ($\theta = 0^\circ$), D ($\theta = 22.5^\circ$) and G ($\theta = 45^\circ$) for polarization configurations LRR-L and RLL-R. (c) Phase distributions, as in (b), after adding or subtracting 4θ phase. Wavelengths of the pump and signal waves are $\lambda_1=1028$ nm and $\lambda_2=1310$ nm, respectively.

4. Conclusion

We have studied the role of rotational symmetry in the four-wave-mixing process from metasurfaces, proved the existence of the geometric Berry phase and its role in FWM. We theoretically and experimentally demonstrated spin-controlled FWM by using plasmonic metasurfaces with four-fold rotational symmetry. We showed that the polarization of FWM can be manipulated by choosing appropriate combination of the polarizations of the excitation beams. At the same time, the intensity of FWM can be controlled by the orientation of the adjacent meta-atoms via a geometric phase. Exploiting the spin-dependent geometric Berry phase can introduce additional degrees of freedom to control FWM on plasmonic and dielectric metasurfaces, and may have important applications in the fields such as phase conjugation, parametric amplification, supercontinuum generation and frequency comb generation.

Supporting Information

Additional supporting information may be found in the online version of this article at the publisher's website.

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Graphical Abstract

The role of rotational symmetry and circular polarized excitation is studied in four-wave mixing processes. It is shown that the four-wave mixing polarization depends on the handedness of the input beams. The influence of the geometric phase on the generated intensity is demonstrated in the nonlinear metasurface.

